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Demonstration of Temporal Distinguishability in a Four-Photon State and a Six-Photon State

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An experiment is performed to demonstrate the temporal distinguishability of a four-photon state and a six-photon state, both from parametric down-conversion. The experiment is based on a multi-photon interference scheme in a recent discovered NOON-state projection measurement. By measuring the visibility of the interference dip, we can distinguish the various scenarios in the temporal distribution of the pairs and thus quantitatively determine the degree of temporal (in)distinguishability of a multi-photon state.

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It has been well-known by now that quantum nonlocality is more dramatic in multi-particle entanglement [1]. It was shown that the amount of locality violation increases with the number of particles [2]. Experimental demonstrations of locality violation have thus been shifted from the traditional test of two-photon Bell's inequalities [3, 4, 5] to the test of generalized Bell's inequalities for three or four photons in various states [6, 7, 8, 9, 10]. While entangled two-photon states are produced naturally from parametric down-conversion, generation of three- and four-photon entangled states has to rely on simultaneous two-pair production in parametric down-conversion. Since pairs are produced randomly in parametric down-conversion process, this raises a question, that is, are the two pairs really in an entangled four-photon state or they are simply independent uncorrelated two pairs?

This question was first attempted by Ou, Rhee, and Wang [11, 12] in an experiment similar to the famous Hong-Ou-Mandel experiment [13] but with two pairs of photons. Recently, a number of experiments were performed to further address the problem of photon pair distinguishability in parametric down-conversion [14, 15, 16, 17]. All the experimental schemes are more or less some sort of multi-photon interference (either two-photon or four-photon). More recently, a new scheme was proposed by Ou [18] that relies on a newly discovered NOON-state projection measurement process [19, 20, 21] to characterize quantitatively the degree of temporal distinguishability of an N-photon state. When it applies to photon pairs from parametric down-conversion, it shows various visibility of multi-photon interference for different scenarios in the temporal distributions of the photons [18].

In this letter, we wish to report on an experimental implementation of the NOON-state projection measurement for characterizing the temporal distinguishability of photon pairs from parametric down-conversion. We find that the temporal distinguishability depends on the visibility of multi-photon interference in NOON state projection. When the pairs are indistinguishable from each other, we obtain the maximum visibility which starts to decrease as the pairs begin to separate from each other and becomes a nonzero minimum when they are well separated and completely distinguishable.

The key idea in Ref.[18] for characterizing temporal distinguishability is the NOON-state projection measurement, as depicted in Fig.1 for $N = 4$ and 6. This was recently proposed and demonstrated by Sun *et al.* [19, 21] and Resch *et al.* [20] for multi-photon de Broglie wavelength. This measurement projects an arbitrary two-mode N-photon state of polarization in the form of

$$|\Psi_N\rangle = \sum_k c_k |N-k\rangle_H |k\rangle_V, \tag{1}$$

to a NOON state of $|NOON\rangle = (|N,0\rangle - |0,N\rangle)/\sqrt{2}$. The outcome of this measurement is proportional to

$$P_N \propto |\langle NOON|\Psi_N\rangle|^2. \tag{2}$$

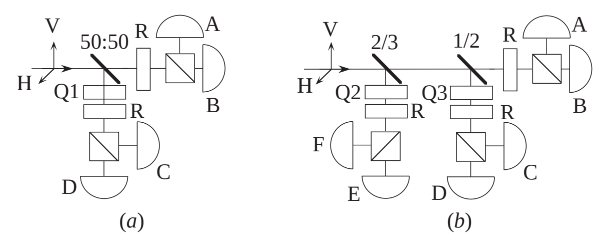

FIG. 1: *A NOON-state projection measurement for (a) four-photon case and (b) six-photon case. Q1 is a phase shifter of $\pi/2$, Q2 of $2\pi/3$, and Q3 of $4\pi/3$. R is a rotator of 45°.*

The physics behind this projection measurement is an ingenious arrangement [22] of beam splitters, phase shifters, and projection polarizers for the cancellation by destructive interference of the middle terms in the form of $|N-k\rangle_H|k\rangle_V$ with $k \neq 0, N$ in Eq.(1). In particular for $|\Psi_4\rangle = |2\rangle_H|2\rangle_V$ and $|\Psi_6\rangle = |3\rangle_H|3\rangle_V$, the projection

probability in Eq.(2) is zero due to orthogonality. These two states are readily available from Type-II parametric down-conversion with a quantum state of

$$|PDC\rangle = |0\rangle + \eta|1\rangle_H|1\rangle_V + \sqrt{2}\eta^2|2\rangle_H|2\rangle_V + \sqrt{6}\eta^3|3\rangle_H|3\rangle_V + ..., \qquad (3)$$

where $|\eta|^2 << 1$ is the pair production probability.

However, the expression in Eq.(2) is for single mode treatment, which means that the four photons or six photons must be in a single temporal mode (the so called 4×1 or 6×1 case). So the pairs must be indistinguishable in the production process. This is normally achieved with an ultra-short pump pulse for parametric down-conversion so that the time of the pair production is restricted in the time duration defined by the pump pulse. But because of the finite duration of the pump pulse, this is only approximately the case in practice. So the pairs actually have partial indistinguishability and this will be reflected in the reduced visibility in the multi-photon interference in the NOON state projection measurement. As suggested in Ref.[18], the visibility is then a direct measure for the degree of indistinguishability.

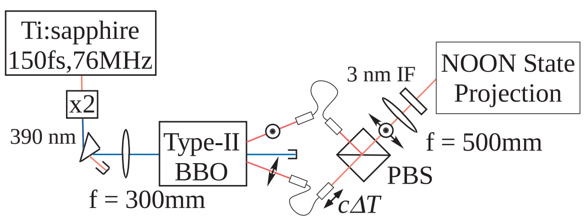

FIG. 2: *Sketch of the experimental set-up*

The experimental arrangement is shown in Fig.2. A 2-mm long BBO crystal cut for Type-II parametric down-conversion is pumped at 390 nm by a frequency-doubled femto-second Ti:sapphire laser. The pump pulse has a width of 150 fsec and a repetition rate of $R_0 = 76$ MHz. The crystal is so oriented that the two conic down-converted fields (o- and e-rays) at the degenerate frequency converge into two unidirectional beams [23]. The two fields are then coupled to single mode polarization preserving optical fibers. The outputs of the fibers are directed to a polarization beam splitter (PBS) to merge into one beam. One of the fiber outputs is mounted on a translational stage so that we can adjust the relative delay $c\Delta T$ between the two polarizations. The recombined beam, after passing through an interference filter of 3 nm width, is sent to the corresponding NOON-state projection measurement assembly for either four- or six-photon case depicted in Fig.1. All combinations of two-photon (say, AB, AC, etc.) and four-photon coincidence (say, ABCD, ACBE, etc.) as well as six-photon coincidence (ABCDEF) in the six-photon case are measured as a function of the relative delay $c\Delta T$ between the two polarizations. Because of the vast amount of coincidence

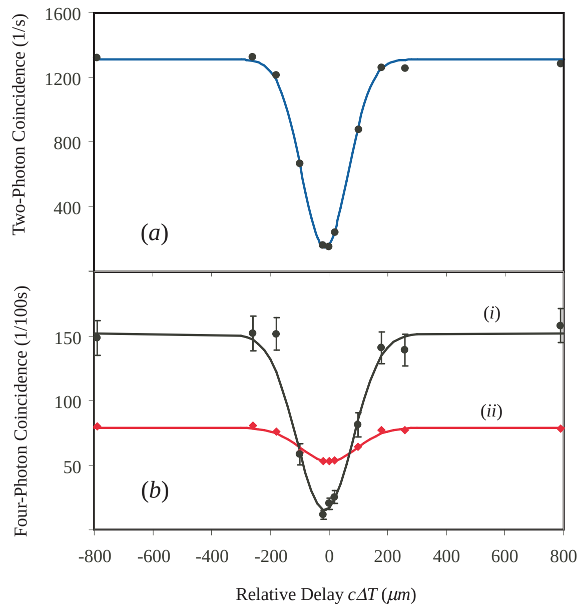

FIG. 3: *Measured two-photon coincidences (a) and four-photon coincidences (b) as a function of relative delay $c\Delta T$. The circles in (b) are direct measured ABCD coincidences while the diamonds are indirectly measured coincidences corresponding to the 2×2 case. The solid curves are Gaussian fit with a visibility of 90% and 33%, respectively.*

data and the lack of coincidence units, we measure each coincidence individually.

Four-photon case. — When four-photon coincidence is measured in the four-photon NOON state projection scheme in Fig.1a, the vacuum and two-photon terms in Eq.(3) make no contribution whereas the six-photon term is higher order (it does produce a background that must be subtracted in data analysis). The four-photon terms can be thought of as two-pair production. From the properties of parametric down-conversion, we know that the two photons within a pair are indistinguishable in time. But since pair production is random, the two pairs are often generated at two well separated times and are described by the quantum state of $|1\rangle_{H1}|1\rangle_{V1} \otimes |1\rangle_{H2}|1\rangle_{V2}$ (the 2×2 case). Or by chance, the two pairs may be generated in the same time and become indistinguishable four photons described by the quantum state of $|2\rangle_H|2\rangle_V$ (the 4×1 case).

For the four-photon NOON-state projection measurement in Fig.1a, there are six possible combinations of two-photon coincidence. They are simply AB, CD, AC, BD, AD, BC. Among them, AB and CD show the typical two-photon Hong-Ou-Mandel dips [13] with AB shown in Fig.3a whereas the rest is flat (not shown). The visibility of the dip is 89%. The directly measured four-photon coincidence ABCD data after background subtraction is shown as solid circles (i) in Fig.3b with a Gaussian fit that has a visibility of 90%. The points in diamonds (ii) are indirectly measured four-photon coinci-

dence data corresponding to the case when the measured four-photon coincidence is from two well-separated pairs of down-converted photons (the 2×2 case). The four-photon coincidence in this case is from pairwise accidental coincidence: there are three possibilities of AB+CD, AC+BD, and AD+BC. So the four-photon coincidence in 2×2 case can be deduced from the measured two-photon coincidences as

$$R_4(ABCD)(2\times 2) = [R_2(AB)R_2(CD) + \\ + R_2(AC)R_2(BD) + R_2(AD)R_2(BC)]/R_0. \quad (4)$$

The solid curve is a Gaussian with a visibility of 33%.

The less than 100% visibility (90%) for the directly measured four-photon coincidence has two origins. One is from imperfect spatial mode match due to misalignment. This has already reduced the two-photon visibility to 89% in Fig.3a. The other origin is from non-overlapping between the two detected pairs of photons. In other word, the two pairs are not completely indistinguishable for us to treat them as in single temporal mode and to use Eq.(2) for four-photon coincidence. Ref.[19] has a complete account of these two effects and derived the visibility under these imperfect conditions as

$$\mathcal{V}_4 = \frac{2v_2(\mathcal{A}+3\mathcal{E}) - v_2^2(\mathcal{A}+\mathcal{E})}{3(\mathcal{A}+\mathcal{E})}. \quad (5)$$

Here v_2 is the two-photon visibility from Fig.3a. $\mathcal{A}$ is proportional to the absolute square of the four-photon wave function whereas $\mathcal{E}(\leq \mathcal{A})$ depends on photon pair exchange symmetry. When $\mathcal{E} = \mathcal{A}$, the two pairs are completely overlapping and the four photons are in an indistinguishable entangled state described by $|2\rangle_H|2\rangle_V$ (the 4×1 case). On the other hand, when $\mathcal{E} = 0$, the two pairs are completely separated from each other and become independent (the 2×2 case) with a four-photon visibility of $\mathcal{V}_4(2\times 2) = 0.33$ from Eq.(5). This value is exactly the value from the diamond points in Fig.3b. However, the direct observed four-photon dip in the circle points in Fig.3b is somewhere in between the two extreme cases. Substituting the observed values of $\mathcal{V}_4 = 0.90$ and $v_2 = 0.89$ in Eq.(5) and solving for $\mathcal{E}/\mathcal{A}$, we obtain $\mathcal{E}/\mathcal{A} = 0.90$. The quantity $\mathcal{E}/\mathcal{A}$ thus provides a measure of partial indistinguishability between the pairs. The nonzero value of 0.33 for $\mathcal{V}_4(2\times 2)$ can be thought of as a result of the indistinguishability between the two photons within each pair. So the directly measured four-photon dip visibility $\mathcal{V}_4$ is a measure of indistinguishability for all the four photons, as suggested in Ref.[18].

$\mathcal{E}/\mathcal{A}$ can be independently measured from two-photon coincidence on one (o- or e-ray) of the the two down-converted fields [11, 12]. This is achieved by blocking one of the two beams that come to the PBS. The directly measured value is $\mathcal{E}/\mathcal{A} = 0.77 \pm 0.06$. Another independent method to measure $\mathcal{E}/\mathcal{A}$ is from the ratio of the values at infinity delay ($|c\Delta T| = \infty$) in the two data sets in Fig.3b. Ref.[19] gives the ratio as $1 + \mathcal{E}/\mathcal{A}$ and from Fig.3b, we find $\mathcal{E}/\mathcal{A} = 0.92$. This value is consistent with the one derived from visibility. However, the value from two-photon coincidence measurement is somewhat smaller than the ones from four-photon measurement. We believe that this is caused by spatial mode match between the two pairs of photons. The four-photon coincidence is more restricted than two-photon coincidence and thus acts as some sort of spatial mode filtering resulting in better mode match and higher $\mathcal{E}/\mathcal{A}$ values.

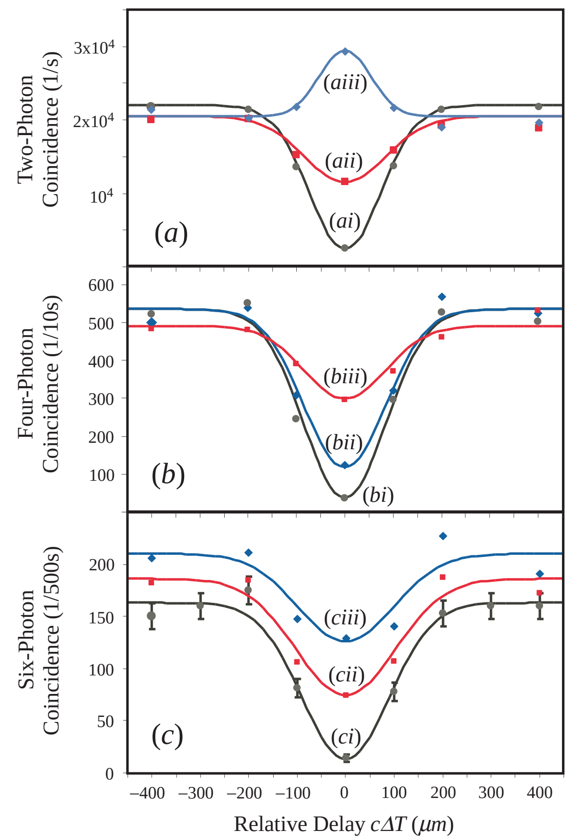

FIG. 4: *Measured two-photon coincidences (a), four-photon coincidences (b), and six-photon coincidences (c) as a function of relative delay $c\Delta T$. (ai) is for AB, (aii) for AE, and (aiii) for BE in (a). (bi) is for ABCE, (bii) for ABCD, and (biii) for BCDE in (b). The solid circles (ci) in (c) are directly measured ABCDEF coincidence while the diamonds (ciii) are indirectly measured coincidence corresponding to 2×3 case and the square points (cii) to the $4\times 1+2$ case. The solid curves are Gaussian fit with a visibility of 92%, 59%, and 39%, respectively. The data in (cii) and (ciii) are multiplied by 2 and 8 respectively to bring them to the same scale as (ci).*

Six-photon case. — With six-photon coincidence, the terms with less than six photons in Eq.(3) have no contribution. Since the six photons are from three pairs of down-converted photons, there are three extreme cases: (1) the three pairs are generated in the same time and are indistinguishable in the quantum state of $|3\rangle_H|3\rangle_V$ (the 6×1 case); (2) two of them are indistinguishable but well separated from the third pair; they are in the quantum

state of $|2\rangle_{H1}|2\rangle_{V1} \otimes |1\rangle_{H2}|1\rangle_{V2}$ (the $4 \times 1 + 2$ case); (3) all three pairs are well separated from each other and are in $|1\rangle_{H1}|1\rangle_{V1} \otimes |1\rangle_{H2}|1\rangle_{V2} \otimes |1\rangle_{H3}|1\rangle_{V3}$ (the 2×3 case). The three cases give three different results in the six-photon NOON-state measurement [18].

In the NOON-state projection for six-photon (Fig.1b), there are 15 different combinations of two-photon or four-photon coincidence. Among the two-photon coincidences, AB, CD, and EF are the same and show a typical Hong-Ou-Mandel dip with 100% visibility in the ideal case; AC, AE, BD, BF, CE, DF are the same with a dip of an ideal 50% visibility; AD, BC, CF, DE, AF, BE are the same with a bump of an ideal 50% visibility. Fig.4a shows AB, AC, and AD. Among the four-photon coincidence, ABCE, ABDF, CDBF, CDAE, EFBD, EFAC are the same with a dip of 100% visibility ideally; ABCF, ABDE, CDBE, CDAF, EFBC, EFAD are the same with a dip of 1/3 visibility ideally; ABCD, ABEF, CDEF are the same with a dip of 5/6 visibility ideally. They are plotted in Fig.4b after background subtraction. The fitted curves give dips with visibility smaller than the ideal ones. The directly measured six-photon coincidence data is presented in Fig.4c as solid circles. The dip in the fitted Gaussian curve has a visibility of 0.92, as compared to the ideal 100%. The diamond points and square points are indirectly measured six-photon coincidence corresponding to the 2×3 (x8) and the $4 \times 1 + 2$ case (x2), respectively. They are deduced from

$$R_6 = \sum_P R_2(P) R_4(P)/R_0, \tag{6}$$

where Ps are the 15 different combinations of (AB) (CDEF). For the $4 \times 1 + 2$ case (square points), the quantities $R_4(CDEF)$, etc. are directly measured four-photon coincidences (shown in Fig.4b) but for the 2×3 case (diamond points), they are derived from two-photon coincidences by using a formula similar to Eq.(4). The diamond points and the square points are fitted to Gaussian functions with visibility of $\mathcal{V}_6(2 \times 3) = 0.39$ and $\mathcal{V}_6(4 \times 1 + 2) = 0.59$, respectively. The ideal values are $\mathcal{V}_6^{(0)}(2 \times 3) = 2/5$ and $\mathcal{V}_6^{(0)}(4 \times 1 + 2) = 3/5$ [18].

The observed visibilities in both 6×1 and $4 \times 1 + 2$ case are close to the ideal values, indicating that the pairs are almost indistinguishable. This is reflected in the directly measured $\mathcal{E}/\mathcal{A} = 0.91 \pm 0.04$ from two-photon coincidence and the $\mathcal{E}/\mathcal{A}$ value would be even closer to 1 for six-photon coincidence. Of course, the observed visibility in 2×3 case is always the predicted value because the two photons in each pair are truly indistinguishable and we have a genuine 2×3 case.

In summary, we used the newly discovered NOON-state projection measurement technique to quantitatively characterize the temporal distinguishability of a four- or six-photon state. We find the visibility of the multi-photon interference can be used to distinguish different scenarios in the temporal distribution of the photons.

Acknowledgments

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